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RADIOIODINE MEASUREMENTS OF THE STACK EFFLUENT FROM THE CP-5 5.0-MW HEAVY-WATER REACTOR

by Weldon D. Dillow

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Industrial Hygiene and Safety Division

February 1968



## TABLE OF CONTENTS

		Page
ABS	STRACT	4
I.	INTRODUCTION	5
II.	COLLECTION EFFICIENCY OF CESCO CARTRIDGE	5
III.	STACK CONCENTRATION MEASUREMENTS	9
IV.	RELATIVE RADIOIODINE RATIOS	11
v.	STACK SINGLE-CHANNEL-ANALYZER RESPONSE	14
VI.	SUMMARY	16
AC	KNOWLEDGMENTS	16
RE:	FERENCES	17

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ABSTRACT

INTRODUCTION

COLLECTION REFLICIENCE OF CREEK CARTRICES

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REFERENCES

# LIST OF FIGURES

No.	Title	Page
1.	CP-5 Ventilation Exhaust System	6
2.	Initial Spectrum of CP-5 Long-term-stack Sample Collected on BPL 12 x 30 Mesh Charcoal-loaded Cartridge from 1315 on 4/12/66 to 0931 on 4/18/66	11
3.	Initial Spectrum of CP-5 Short-term-stack Sample Collected on CESCO Cartridge from 1000-1315 on $3/10/66$	11
4.	Five-day Decay Spectrum of CP-5 Short-term-stack Sample Collected on CESCO Cartridge from 1000-1315 on $3/10/66$	12
5.	Spectrum of CP-5 Long-term-stack Sample Counted with a Ge(Li) Detector Collected from 1426 on 5/2/66 to 0837 on 5/9/66	12
6.	Relative Radioiodine Activity as a Function of Sampling Time $$ .	14
7.	CESCO Cartridge in Place in the Detector Shield	14
8.	CP-5 Stack SCA <sup>131</sup> I Counting Rate as a Function of Sampling Time	16
	LIST OF TABLES	
No.	<u>Title</u>	Page
I.	Distribution of 131 Activity by Cartridge	7
II.	131 I Concentration Measurements	8
III.	131 I and 133 I Concentrations and Ratios of 133 I to 131 I in CP-5 Stack Effluent	10
IV.	Conditions	10
v.	Ratios of Radioiodine Concentrations in CP-5 Stack Effluent Relative to <sup>131</sup> I	13
VI.	131 I Contribution to the Initial Count Obtained with a 512-channel Analyzer	15

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# RADIOIODINE MEASUREMENTS OF THE STACK EFFLUENT FROM THE CP-5 5.0-MW HEAVY-WATER REACTOR

by

Weldon D. Dillow

### ABSTRACT

This report describes experimental measurements and analyses of the radioiodines in the stack effluent of the CP-5 Reactor.

The objective of this work was the acquisition of data essential to analyzing the response of the present CP-5 iodine stack monitor and to determining the requirements of other permanent iodine monitors to be installed at various site locations. The scope of the investigation consisted of making the following measurements and analyses: (1) radioiodine collection efficiency of the Chicago Eye Shield Company's charcoal-loaded organic vapor respirator cartridge (CESCO); (2) average <sup>131</sup>I and <sup>133</sup>I concentrations; (3) ratios of <sup>132</sup>I, <sup>133</sup>I, <sup>134</sup>I, and <sup>135</sup>I relative to <sup>131</sup>I, based on analyses of scrubber samples; and (4) analysis of the response of the present single-channel-analyzer (SCA) stack-monitoring system to a complex radioiodine spectrum.

Results indicated that the CESCO cartridge is a satisfactory radio-iodine collector with a collection efficiency of 92%. Average  $^{131}I$  and  $^{133}I$  stack concentrations ranged from 2.00 to 3.69 x  $10^{-10}$  and 2.01 to 2.72 x  $10^{-9}~\mu\text{Ci/cm}^3$ , respectively. In only one of the four scrubber samples was  $^{132}I$  positively identified. The  $^{133}I/^{131}I$  ratios for operation at full reactor power determined from the scrubber samples ranged from 6.3 to 10.1, and were consistent with the ratios determined by charcoal-sampling techniques. The  $^{134}I/^{131}I$  and  $^{135}I/^{131}I$  ratios were more variable, varying by factors of three and two, respectively. Finally, the single-channel-analyzer response to  $^{131}I$  was not singular, but was a multiple response to the other radioiodines as well as a result of their Compton continuum contributions to the  $^{131}I$  photopeak. The  $^{131}I$  contribution to the total count seen in the window of the single-channel analyzer was primarily a function of the sampling time, and could not be quantitatively determined during sample collection.

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Results indirected that the CESCO caircidge is a satisfactory radional collector with a collection efficiency of 925. Average "1; and "2; all stack acceptants from 2.00 to 1.00 x [97] and 2.00 at 2.20 and 2.01 to 2.72 and 1.00 and 2.00 a

#### I. INTRODUCTION

Measurements to determine the radioiodine content of the CP-5 Reactor's stack effluent were made for the following purposes:

- (1) To understand the response of the present CP-5 iodine-monitoring system to a complex radioiodine spectrum,
- (2) To provide basic data to be used in establishing the requirements of other permanent iodine monitors to be installed at various site locations. Other iodine monitors refer to one under development by the Laboratory's Industrial Hygiene and Safety (IHS) Division and to commercial equipment being considered as part of new facilities.

The scope of this investigation included the following measurements: (1) radioiodine collection efficiency of the IHS-stocked CESCO\* charcoal cartridges used in most <sup>131</sup>I monitoring applications; (2) average stack <sup>131</sup>I and <sup>133</sup>I concentrations; and (3) ratios of <sup>132</sup>I, <sup>133</sup>I, <sup>134</sup>I, and <sup>135</sup>I relative to <sup>131</sup>I, according to analyses of scrubber samples. Based on the above measurements, the response of the present stack monitor (a single-channel analyzer set on the 364-keV <sup>131</sup>I photopeak) was analyzed.

All iodine samples were collected from the fan-room plenum, as shown by the sampler location in Fig. 1. The most likely sources of radio-activity during normal operation, i.e., the thimble-cooling and experimental beam-hole ventilating systems, were filtered by high-efficiency particulate filters. In all cases, the iodine sampler was preceded by a HV-70 filter to eliminate particulate aerosol activity.

## II. COLLECTION EFFICIENCY OF CESCO CARTRDIGE

The first item investigated was the iodine-collection efficiency of the fixed-filter charcoal cartridge used as the collector in the prototype monitor. This charcoal cartridge, designated CESCO, is manufactured for use as a respirator filter to provide protection against organic vapors. Contact with the manufacturer yielded the sketchy information that the charcoal in the cartridge is a commercial-grade 16 x 16-mesh size produced by Pittsburgh Activated Carbon Company.

In some measurements, as a check on the effectiveness of the CESCO cartridge, Pittsburgh Activated Carbon Company's 12 x 30-mesh BPL granular activated carbon was substituted for the CESCO charcoal.

For these investigations, three cartridges were usually connected in series. The series combination was assumed to be 100% efficient, although

<sup>\*</sup>Chicago Eye Shield Company Safety Products, Inc., 2727 W. Roscoe St., Chicago, Ill.

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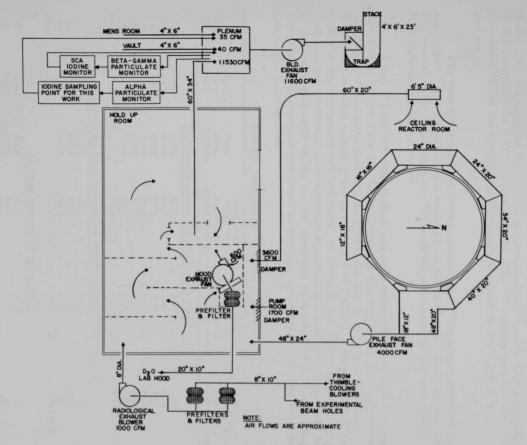


Fig. 1. CP-5 Ventilation Exhaust System



this assumption is not necessarily so since the presence of radioiodine in the third cartridge implies that some probably penetrated that cartridge. After sufficient time had elapsed to let the shorter-lived radioiodines <sup>132</sup>I, <sup>134</sup>I, and <sup>135</sup>I decay, the cartridges were counted with a Nuclear Data Model 130, 512-channel analyzer to determine the <sup>133</sup>I content by summing up the counts in the 0.530-keV photopeak. The <sup>131</sup>I content was determined after the <sup>133</sup>I had decayed by summing up the counts in the 0.364-keV photopeak.

The counting efficiency of the analyzer for  $^{131}$ I was measured using a standard  $^{131}$ I solution\* with a reported accuracy of  $\pm 3\%$ . Since no standard was available for measuring the  $^{133}$ I counting efficiency, it was calculated from data in the literature.  $^{1,2}$ 

Total <sup>131</sup>I activity distribution and collection efficiency data are summarized in Tables I and II. Table I lists the distribution of total activity by cartridge for sampling times of less than one day to as long as 7.8 days. Based on the mean values given in column 4 of Table I, a single CESCO cartridge is approximately 92% efficient. In Table II, the collection efficiency of the second cartridge is determined in terms of the activity presented to it; that is, the activity on the second cartridge is divided by the sum of the activities on the second and third cartridges, again assuming total absorption in the combination.

TABLE I. Distribution of 131 I Activity by Cartridge

Run No.	Sampling Time, days	Flow Rate, m <sup>3</sup> /min	Percent of Total <sup>131</sup> I Activity on First Cartridge	Percent of Total <sup>131</sup> I Activity on Second Cartridge	Percent of Total <sup>131</sup> I Activity on Third Cartridge
	mm 1/9 - 63	All Three C	artridges BPL 12 x 3	0 Mesh Charcoal	The same of the last
4	0.96	0.240	95.6	2.48	1.74
5	0.95	0.240	97.7	1.49	0.78
6	2.95	0.230	97.3	2.08	0.61
7	3.88	0.230	98.4	1.28	0.32
8	3,00	0.210	97.3	2.17	0.48
9	5.85	0.216	94.9	3.04	2.04
Mean			96.9 ± 1.33	2.09 ± 0.64	0.995 ± 0.72
	First Ca	artridge CESC	O; Second and Third	BPL 12 x 30 Mesh Char	rcoal
10	4.93	0.219	92.5	6.00	1.48
11	5.98	0.210	92.2	6,73	1.10
12	6.76	0.228	90.6	8.19	1.24
Mean			91.8 ± 1.02	6.97 ± 1.12	1.27 ± 0.19
		All T	nree Cartridges Regu	lar CESCO	
17	3.99	0.235	91.9	6.84	1.21
19	7.82	0.242	92.8	5.71	1.53
	6.99	0.245	89.6	8.90	1.62
20					1.02
	1,14	0.231	95.2	3.78	1.04
20		0.231 0.222	95.2 92.2	3.78 6.65	

the third carriage implies that some probably penetrated that carridge After aufficient time had elapsed to let the shorter-lived radioiodines the fit and the decay the carridges were counted with a Nuclear Data Model, 130, 512-channel analyses to determine the the the fill centent by summany up the counts in the 0.530-keV photograk. The 'I'l sentent was deformined after the the the the the country of the co

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TABLE II. Charcoal Cartridge 131 Collection Efficiency and CP-5 Stack 131 Concentration Measurements

Run No.	Type of Charcoal	Sampling Time, days	Residence Time per Cartridge, sec	Collection Efficiency of First Cartridge,	Collection Efficiency of Second Cartridge,	Average 131 Concentration over Sampling Period, µCi/cm <sup>3</sup>	Remarks
1	CESCO	9.96	0.016	90.1		2.19 x 10 <sup>-10</sup>	Primary cartridge was left in place in the prototype
1A 1B 1C	CESCO CESCO CESCO	0.86 0.29 3.97	0.016 0.016 0.016		86.4 80.0 83.3	2.19 x 10 <sup>-10</sup> 2.19 x 10 <sup>-10</sup> 2.19 x 10 <sup>-10</sup>	monitor for approximately 10 days.
2	CESCO	0.83	0.016	97.2		3.34 x 10 <sup>-10</sup>	No third cartridge, so efficiency of second could not be determined.
3	No. 1 Silica Gel; Nos. 2 and 3 CESCO	0.94	0.015	34.9	95.3	3.69 x 10 <sup>-10</sup>	First cartridge was loaded with silica gel.
4	BPL	0.96	0.016	95.6	58.8	2.46 x 10 <sup>-10</sup>	All three cartridges were loaded with 8PL 12 x 30 mesh
5	BPL	0.95	0.016	97.7	65.7	2.47 x 10 <sup>-10</sup>	All three cartridges were loaded with BPL 12 x 30 mesh
6	BPL	2.95	0.017	97.3	77.2	2.55 x 10 <sup>-10</sup>	All three cartridges were loaded with BPL 12 x 30 mesh
7	BPL	3.88	0.017	98.4	79.9	2.37 x 10 <sup>-10</sup>	All three cartridges were loaded with BPL 12 x 30 mesh
8	BPL	3.00	0.019	97.3	81.7	3.18 x 10 <sup>-10</sup>	All three cartridges were loaded with BPL 12 x 30 mesh
9	BPL	5.85	0.018	94.9	59.9	3.31 x 10 <sup>-10</sup>	All three cartridges were loaded with BPL 12 x 30 mesh
10	CESCO-BPL	4.93	0.019	92.5	80.1	1.70 x 10 <sup>-10</sup>	Cartridge 1 was a CESCO. Cartridges 2 and 3 were loaded with BPL 12 x 30 mesh. Reactor was down 2.14 days before end of sampling period.
11	CESCO-BPL	5.98	0.019	92.2	86,0	2.09 x 10 <sup>-10</sup>	Cartridge 1 was a CESCO. Cartridges 2 and 3 were loaded with BPL 12 x 30 mesh.
12	CESCO-BPL	6.76	0.017	90.6	86.8	2.63 x 10 <sup>-10</sup>	Cartridge 1 was a CESCO. Cartridges 2 and 3 were loaded with BPL 12 x 30 mesh.
13	CESCO-BPL	6.91	0.020	94.6	79.5	2.66 x 10 <sup>-10</sup>	Cartridge 1 was a CESCO. Cartridges 2 and 3 were loaded with BPL 12 x 30 mesh. Reactor was down 8.0 h before end of sampling period.
14	CESCO	0.88	0.013	named a	puir incinc	0.51 x 10 <sup>-10</sup>	Only cartridge 1, which was a CESCO, was used. Reactor was down during entire sampling period. A collection efficiency of 92.5% was assumed (based on the average or runs 10-13).
15	CESCO	0.97	0.013	Sant Go	the last tenden	0.41 x 10 <sup>-10</sup>	Reactor was down during entire sampling period. Same remarks apply as to run 14 above.
16	CESCO	1.00	0.013	-	-	2.09 x 10 <sup>-10</sup>	Only cartridge 1, which was a CESCO, was used. A collection efficiency of 92,5% was assumed.
17	CESCO	3.99	0.017	91.9	85.0	2.54 x 10 <sup>-10</sup>	All three cartridges were CESCO.
18	CESCO	0.98	0.014	in cha	roja:	0.72 x 10 <sup>-10</sup>	Only cartridge 1, which was a CESCO, was used. A col- lection efficiency of 92.5% was assumed. Reactor was down during entire sampling period.
19	CESCO	7.82	0.016	92.8	73.8	2.72 x 10 <sup>-10</sup>	All three cartridges were CESCO.
20	CESCO	6.99	0.016	89.6	84,5	2.96 x 10 <sup>-10</sup>	All three cartridges were CESCO.
21	CESCO	1.14	0.017	95.2	78.4	0.49 x 10 <sup>-10</sup>	All three cartridges were CESCO. Reactor was down during entire sampling period.
22	CESCO	7.80	0.018	92.2	84.9	2.00 x 10 <sup>-10</sup>	All three cartridges were CESCO.

The mean collection efficiency of the second cartridge was  $78.4\pm8.3\%$ . Data from run No. 3 were discarded since the first cartridge was loaded with silica gel, which was only 34.9% efficient. The collection efficiency of the third cartridge could not be determined since there were no collecting media downstream from it. Why the second cartridge was 14-18% less efficient than the first cartridge was not definitely established, but it may have resulted from the presence of a much more penetrating organic iodide. Several investigators have identified methyl iodide as a small component (usually less than 5% of the total iodine content<sup>3</sup>) of fission-productiodine-carrying gas streams.

Results of the above collection-efficiency measurements should not be applied generally across the board to other facilities without confirmatory data. Since most charcoals have been nearly 100% effective for elemental iodine, the real problem in radioiodine removal arises from the a residu at in and side mending will are the ballega and

penetrating forms that may be present in varying percentages from facility to facility. Therefore, unimpregnated charcoals require in-place confirmatory test data at frequent intervals.

## III. STACK CONCENTRATION MEASUREMENTS

The average <sup>131</sup>I and <sup>133</sup>I concentration measurements in the CP-5 stack effluent are presented in Tables II and III, respectively. These average concentrations were calculated by using the following equation:

$$C_{i} = \frac{\lambda_{i}A_{i} \exp(\lambda_{i}t_{d})}{F_{g_{i}}Ck[1 - \exp(-\lambda_{i}t_{s})]}$$
(1)

where

C<sub>i</sub> = Concentration of ith isotope, in  $\mu$ Ci/cm<sup>3</sup>;

 $\lambda_i$  = Decay constant of ith isotope, in min<sup>-1</sup>;

A<sub>i</sub> = Number of counts per minute on the first cartridge in the respective photopeak for which the counter efficiency has been measured or calculated;

t<sub>d</sub> = Decay time, in minutes, since the end of the sampling period;

F = Flow rate, in cm<sup>3</sup>/min;

g<sub>i</sub> = Counter efficiency for the iodine photopeak under which summation is made (measured to be 8.85% for <sup>131</sup>I; calculated to be 8.14% for <sup>133</sup>I);

C = Fractional collection efficiency of the first cartridge;

 $k = 2.22 \times 10^6 \, dpm/\mu Ci;$ 

and

t<sub>s</sub> = Sampling time, in minutes.

If F is expressed in  $m^3/min$ , and  $A_{I-131}$  and  $A_{I-133}$  represent the number of counts/min less the estimated background and Compton scattering in channels 32-40 and 48-58 for  $^{131}I$  and  $^{133}I$ , respectively, the equations for calculating the  $^{131}I$  and  $^{133}I$  concentrations are

$$C_{I-131} = \frac{3.04 \times 10^{-16} A_{I-131} \exp(5.96 \times 10^{-5} t_d)}{FC[1 - \exp(-5.96 \times 10^{-5})(t_s)]}$$
(2)

and

$$C_{I-133} = \frac{3.07 \times 10^{-15} A_{I-133} \exp(5.55 \times 10^{-4} t_d)}{FC[1 - \exp(-5.55 \times 10^{-4} t_s)]}.$$
 (3)

TABLE III. 131 and 133 Concentrations and Ratios of 133 I to 131 in CP-5 Stack Effluent

Run No.	Sampling Time, days	Average <sup>131</sup> I Concentration over Sampling Period, µCi/cm <sup>3</sup>	Average <sup>133</sup> I Concentration over Sampling Period, #Ci/cm <sup>3</sup>	133 <sub>I</sub> /131 <sub>I</sub> Ratio	Remarks
9	5.85	3.31 x 10 <sup>-10</sup>	2.72 x 10 <sup>-9</sup>	8.2	These the marking of the
11	5.98	2.09 x 10 <sup>-10</sup>	2.39 x 10 <sup>-9</sup>	11.4	
12	6.76	$2.63 \times 10^{-10}$	2.01 x 10 <sup>-9</sup>	7.6	
13	6.91	2.66 x 10 <sup>-10</sup>	1.96 x 10 <sup>-9</sup>	7.4	Reactor was shut down 9 hr before end of sampling.
14	0.88	$0.51 \times 10^{-10}$	0.23 x 10 <sup>-9</sup>	4.5	Reactor was shut down during entire sampling period.
15	0.97	$0.41 \times 10^{-10}$	0.34 x 10 <sup>-9</sup>	8.4	Reactor was shut down during entire sampling period.
16	1.00	$2.09 \times 10^{-10}$	1.39 x 10 <sup>-9</sup>	6.6	Sampling began 3 hr before reactor startup.
17	3.99	$2.54 \times 10^{-10}$	2.39 x 10 <sup>-9</sup>	9.4	
18	0.98	$0.72 \times 10^{-10}$	0.34 x 10 <sup>-9</sup>	4.7	Reactor was shut down during entire sampling period.
19	7.82	$2.72 \times 10^{-10}$	2.62 x 10 <sup>-9</sup>	9.6	
22	7.80	$2.00 \times 10^{-10}$	2.45 x 10 <sup>-9</sup>	12.2	

It can be seen from column 7 of Table II that, with the reactor operating at 5.0 MW, the  $^{131}I$  concentration ranged from 2.00 to 3.69 x  $10^{-10}\,\mu\text{Ci/cm}^3$  and was generally independent of the sampling time. When the reactor was shut down, the  $^{131}I$  concentration was one-fifth to one-fourth of that measured during normal operation and time dependent.

Neglecting those cases in Table III in which the reactor was not at full power during the sampling period, the data show that the <sup>133</sup>I concen-

TABLE IV. Ratios of <sup>133</sup>I to <sup>131</sup>I Activity for Fresh and Equilibrium Conditions

	133I/131I Ratio			
Decay Time	Fresh Condition	Equilibrium Condition		
0	21	2.3		
l hr	19	2.2		
l day	9.9	1.1		
2 days	4.9	0.55		
3 days	-	0.27		
4 days	5 100 - 100 -	0.13		

tration ranged from 2.01 to 2.72 x  $10^{-9} \, \mu \text{Ci/cm}^3$ . When the reactor was down, the <sup>133</sup>I concentration was reduced one-tenth to one-eighth of its value at full power. The <sup>133</sup>I/<sup>131</sup>I ratio at power ranged from 7.6 to 12.2. Table IV<sup>4</sup> shows the <sup>133</sup>I/<sup>131</sup>I activity ratios in fresh and equilibrium fission products at various decay intervals. Thus, the ratios found in the CP-5 stack effluent were indicative of one-day-old fresh fission products.

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Is the be seen from Column 5 of Table 15 that which where open a simple search of the column open a sing at 5.0 MW, the "" concentration require the column to the column to the column that was generally independent as the above the concentration was one-ship to interference that negative the concentration was one-ship to interference that the negative the concentration and time dependence.

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## IV. RELATIVE RADIOIODINE RATIOS

It became apparent from observations of the 512-channel analyzer spectrum that the quantitative determination of the individual iodine components could not be accomplished simply by gamma analyses. This can be seen from Fig. 2, which shows the <sup>131</sup>I and <sup>133</sup>I photopeaks clearly, but does not permit resolution of the higher-energy photopeaks. Also, the higher-energy components produce a Compton continuum, which contributes counts to the <sup>131</sup>I and <sup>133</sup>I photopeaks. This is shown by Figs. 3 and 4, which are the spectra for a short-term sample counted immediately and five days after the end of collection, respectively. Note the masking of the <sup>131</sup>I photopeak in the first count. One of the cartridges was counted with a Ge(Li) detector in an attempt at resolution of the shorter-lived radioiodines, but this did not prove satisfactory either. Figure 5 shows the spectrum obtained with the Ge(Li) detector. Note that the <sup>131</sup>I and <sup>133</sup>I photopeaks are prominent, but the higher-energey components are basically swallowed up in the Compton continuum.

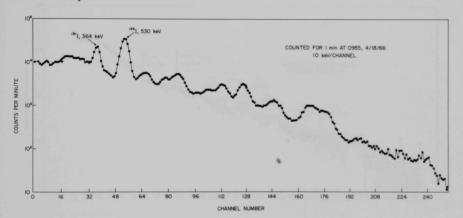


Fig. 2. Initial Spectrum of CP-5 Long-term-stack Sample Collected on BPL 12 x 30 Mesh Charcoal-loaded Cartridge from 1315 on 4/12/66 to 0931 on 4/18/66

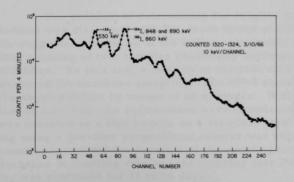


Fig. 3 Initial Spectrum of CP-5 Shortterm-stack Sample Collected on CESCO Cartridge from 1000-1315 on 3/10/66

#### REPRESENTATION RADIO OF THE RATIOS

If became apparent from observations of the Siz-channel analyses appertrum that the quantitalive determination of the individual indine came powerful could not be accomplished simply by gamma analyses. This can be seen from Fig. 4, which shows the Mil and Mil photopeaks. Also, the Mose not permit resolution of the higher-whency photopeaks. Also, the Migher-energy components produce a Compton combinuous, which controlled which are the special to the Mil photopeaks. This is shown by Figs. 3 and a which are the special for a short term sample counted immediately and which are the special for a short term sample counted immediately and Mil photopeak in the first comb. One of the cartestops was counted with the Collision for the state of the state of the special objective of the but this did not prove satisfactory sitler. Figure 5 shows the special objective as taking with the Cellis detector. Mose that the Mil and Mil photopeaks are prominent, but the higher-energy components are basically swallowed as growninent, but the higher-energy components are basically swallowed as



Fig. 2. Edited Spectrum of CP-5 Long-nemi-cated decayte Colleges; of 90, 17 x to stall a Charcoat-loaded Carnidge from 1935 an 4/13/45 on 4/15 on 4/15/55



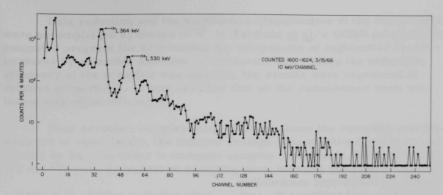


Fig. 4. Five-day Spectrum of CP-5 Short-term-stack Sample Collected on CESCO Cartridge from 1000-1315 on 3/10/66

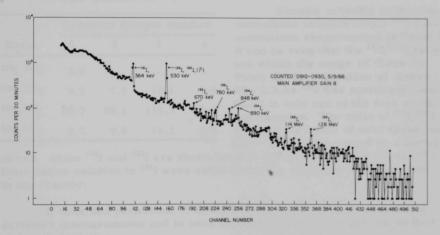


Fig. 5. Spectrum of CP-5 Long-term-stack Sample Counted with a Ge(Li) Detector Collected from 1426 on 5/2/66 to 0837 on 5/9/66

The problem of determining the relative proportions of the radioiodines was discussed with Dr. Jacob Sedlet of the IHS Background Group.
He suggested resolution of the iodine components by extended periods of
beta counting of thin samples chemically separated from scrubber-sampling
solutions. To this end, a one-molar solution each of NaOH and NaHSO3 was
prepared for scrubber sampling. The CP-5 stack effluent was sampled with
the above solution by means of an impinger. Within 45 min of the completion of sampling, the radioiodines were chemically separated, the precipitate
was mounted as a thin sample on a stainless steel planchet, and beta counting using a gas-flow proportional counter was begun. The interval between
counting times and the duration of the counting times were adjusted to the
decrease in counting rate and the counting rate itself, respectively. Counting was continued for 15 to 18 days.



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Data reduction and the quantitative identification of the components were accomplished by means of W. D. Fairman et al.'s CORD program,<sup>5</sup> a computer program for determining the components of radioactive decay mixtures by computer analyses of count-rate data. Since the collection efficiency of the scrubber was unknown, the results were expressed in relative proportions. It was assumed that all the radioiodines were collected with equal efficiency.

Four scrubber samples were secured. Since the sampling periods were not of equal length, the relative average concentrations had to be calculated by Eq. 1 in order to compare samples. Otherwise, the amounts of <sup>131</sup>I and <sup>133</sup>I were dependent on sampling time, whereas the shorter-half-life

TABLE V. Ratios of Radioiodine Concentrations in CP-5 Stack Effluent Relative to <sup>131</sup>I

	Scrub	ber San	mple Nu	mber
Ratio	1	2	3	4
<sup>132</sup> I/ <sup>131</sup> I	3.4		-	-
$^{133}I/^{131}I$	6.3	7.3	10.1	6.5
134 <sub>I</sub> /131 <sub>I</sub>	85.7	78.3	119.3	36.4
135 <sub>I</sub> /131 <sub>I</sub>	9.7	9.0	16.3	8.7

radioiodines approached an independence of sampling time with the approach of sampling equilibrium.

Results in terms of the concentrations relative to the  $^{131}$ I concentrations are presented in Table V. It can be seen that the  $^{133}$ I/ $^{131}$ I ratios are within the range of those in Table III for operation at power. The 2.3-hr  $^{132}$ I was positively identified in only one of the four scrubber samples. The ratio of  $^{134}$ I/ $^{131}$ I varied by a factor of over three, and the  $^{135}$ I/ $^{131}$ I ratio varied by a factor

of two. Since  $^{134}I$  and  $^{135}I$  are short-lived in comparison with 8.07-day  $^{131}I$ , their ratios relative to  $^{131}I$  were sensitive to the variation of holdup time in the reactor.

If time had permitted, it would have been desirable to repeat the scrubber measurements and to have followed the decay by gamma as well as beta counting.

Significant possible sources of error in the scrubber analyses were the determination of the counter yield, which was a sensitive function of beta energy and sample thickness, and the in-growth of the 5.27-day <sup>133</sup>Xe and 9.2-hr <sup>135</sup>Xe daughters, which may have contributed to measured activities. In the CORD program, the daughters are assumed to remain with the parent. However, in the above measurements, it is not unreasonable to assume that some of the xenon atoms diffused from the sample and were swept away by the counting gas.

Using the average of the four ratio determinations listed in Table V, the relative activity of <sup>131</sup>I, <sup>132</sup>I, <sup>133</sup>I, <sup>134</sup>I, and <sup>135</sup>I is plotted in Fig. 6. Note

Data reduction and the quantitative identify all on the components were accomplished by means of W. Dell'alizman at all a CORD program," a component program for determining the components of radicartive decay minimizes by computer scalings of spend-rate data. Since the collection cliffware of the accidible was unsudent the results were expressed in relative proportions. It was assumed that all the radiciones were collected with equal afficiency.

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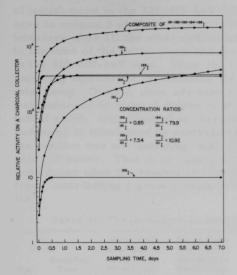


Fig. 6. Relative Radioiodine Activity as a Function of Sampling Time

that after the end of the first and second day's sampling, the composite radioiodine activity has reached 62.5 and 75%, respectively, of its seven-day value.

# V. STACK SINGLE-CHANNEL-ANALYZER RESPONSE

It seems appropriate to discuss the response of the single-channel analyzer (SCA) that is presently installed as a stack <sup>131</sup>I monitor to the complex radioiodine spectrum characteristic of the CP-5 effluent shown in Fig. 2. Briefly described, the monitor consists of an ANL-fabricated single-channel analyzer equipped with a 2 x 2-in. NaI(T1) detector, which constantly views a CESCO cartridge. Since the monitor has been installed to detect <sup>131</sup>I, the

SCA window level is set at 364 keV and the window width at 2%. To avoid interference from gamma-emitting particulates, the monitor is located downstream of a particulate monitor, as shown in Fig. 1. The shield and the CESCO cartridge collector are shown in Fig. 7.

As mentioned previously, the other radioiodines (132I, 133I, 134I, and 135I with gamma rays more energetic than the principal 364-keV gamma from 131I) contribute to the window counting rate of the latter by Compton interaction. This is specifically illustrated in Fig. 3, which is a 512-channel analyzer gamma spectrum of a 3.25-hr sample obtained approximately 10 min from the cessation of sampling. No peak is apparent in the channel 36-37 region corresponding to the 364-keV 131I photopeak. However, Fig. 4 shows the same sample five days later with



Fig. 7. CESCO Cartridge in Place in the Detector Shield

a clearly identifiable <sup>131</sup>I photopeak in the channel 36-37 region. Further illustration of the Compton continuum contribution is given by the multichannel analyzer data presented in Table VI, which gives the percentages

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of initial count in the 320-400-keV span contributed by <sup>131</sup>I. The percentages in column 5 of Table VI were computed by correcting the <sup>131</sup>I count (obtained after all the other radioiodines had decayed to negligible values) to the time of the initial count and dividing this value by the initial count in the 320-400-keV span. Thus, the longer the sampling period (until <sup>131</sup>I sampling equilibrium is reached), the larger the <sup>131</sup>I contribution to the total count. Of note, also, are runs 8 and 17 in which the <sup>131</sup>I contribution was enhanced because of delays of 48.7 and 26.4 hr, respectively, in the initial count. The effect of an increased <sup>131</sup>I contribution during reactor downtime is illustrated by contrasting runs 14, 15, and 18, obtained when the reactor was not operating, with runs 3, 4, and 5, obtained with the reactor at power. This is an expected result since the short-lived isotopes soon decay after the reactor is shut down, leaving the longer-lived isotopes contributing a great portion to the total activity at any given counting time.

TABLE VI. 131 Contribution to the Initial Count Obtained with a 512-channel Analyzer

Run No.	Sampling Time, days	Decay Time from End of Sampling to First Count, hr	Decay Time from End of Sampling to Final Count, days	Percent of Initial Count due to <sup>131</sup> I	Remarks
1	9.66	2.6	23.11	86.8	
1A	0.86	0.5	23.11	2.33	
1B	0.29	0.5	22.83	9.48	
1C	3.97	2.3	23.13	64.7	
2	0.83	0.6	22.72	19.2	
3	0.94	1.3	21.18	26.8	
4	0.96	4.4	20.13	31.7	
5	0.95	0.65	19.20	21.7	
6	2.95	0.30	16.21	46.5	
7	3.88	1.6	12.11	49.1	
8	3.00	48.7	9.09	86.8	
9	5.85	3.5	14.12	44.8	
10	4.93	1.5	11.16	82.7	Reactor was down 2.14 days before end of sampling period.
11	5.98	3.5	15.06	63.6	
12	6.76	4.5	8.08	76.7	
13	6.91	0.6	4.07	69.4	Reactor was down 8.0 hr before end of sampling period.
14	0.88	0.5	7.03	45.1	Reactor was down during entire sampling period.
15	0.97	0.25	13.12	53.4	Reactor was down during entire sampling period.
16	1.00	0.75	12.10	26.2	
17	3.99	26.40	8.12	79.2	
18	0.98	1.00	7.13	50.5	Reactor was down during entire sampling period.
19	7.82	2.1	7.06	64.3	
20	6.99	2.1	9.06	68.7	
21	1.14	0.6	7.91	16.3	Reactor was down and under test at low power.
22	7.80	1.7	20.25	60.8	

Using the data of Tables I and II, the measured detector yield of 1.2%, and an estimated flow rate of 3 cfm, we can compute the CP-5 SCA monitor counting rate from  $^{131}$ I from Eq. 1. If it is assumed that the

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Carry the date of Tables 1 and II, the measured detector yield of 1.7%, and an extracted flow rate of 3 clm, we can compute the CP-5 SCA monitor founding rate from <sup>12</sup>1 from Eq. ? If it is assumed that the

flow rate is constant at 3 cfm, the collection efficiency is 92%, and the stack  $^{131}$ I concentration is 2.5 x  $10^{-10} \,\mu\text{Ci/cm}^3$  (a value fairly representative of Table II values), counting rate may be calculated as a function of

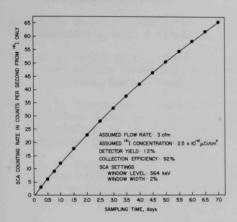


Fig. 8. CP-5 Stack SCA <sup>131</sup>I Counting Rate as a Function of Sampling Time

time. The results of these calculations are presented in Fig. 8.

#### VI. SUMMARY

Iodine isotopes consisting of <sup>131</sup>I, <sup>133</sup>I, <sup>134</sup>I, and <sup>135</sup>I have been identified in the CP-5 stack effluent, and their ratios determined relative to <sup>131</sup>I. In only one of four scrubber samples was <sup>132</sup>I positively identified. The <sup>133</sup>I/<sup>131</sup>I ratios as determined from the analyses of scrubber samples were for the most part within the range determined by charcoalsampling methods, although the latter used much longer sampling times.

Concentrations of  $^{131}I$  and  $^{133}I$  ranged from 2.00 to 3.69 x  $10^{-10}$  and 2.01 to 2.72 x  $10^{-9}~\mu \text{Ci/cm}^3$ , respectively. The  $^{131}I$  present on the cartridge could not be determined quantitatively from the counting rate in the  $^{131}I$  window of the single-channel-analyzer stack monitor because of interference from other radioiodines collected on the cartridge with equal proficiency. The CESCO cartridge collection efficiency averaged 92% for sampling times of one to eight days and is considered acceptable for CP-5 stack-monitoring application. This statement may not apply equally well in other facilities where the chemical makeup of the radioiodine content may be different because of the presence of organic iodides, especially methyl iodide. Therefore, wherever the CESCO cartrdige is to be used, its collection efficiency should be determined by in-place testing using three cartridges in series, as described in this report.

#### ACKNOWLEDGMENTS

I wish to express my thanks to the many people who contributed to this work. Among them are Dr. Jacob Sedlet and his staff, for suggestions on scrubber sampling and assistance in the radiochemistry and counting; W. D. Fairman, for analyzing the scrubber-sample counting data by means of his CORD program; A. G. Januska and his staff, for assistance in counting the scrubber samples; and Andris Peterson, U.S. Public Health Fellow, for his aid in some of the sample collection, radiochemistry, and counting.

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# VI. SUMMARHY

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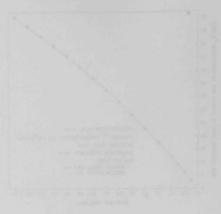


Fig. 8. CF-5 stack SCA 15I Counting Surese a Function of Sampling Tune-

Concentrations of "I and "I ranged from 2.00.10' 3.60 x 10 " and 2.01 to 2.72 x 10" and 2.01 to 2.

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W. D. Fairman, for analyzing the acribber-sample counting only the CORD program; A. C. Janti, a and his staff, for assistance in counting the scrubber samples; and Andria Pererson U.S. Public Health Fellow, for his aid in some of the sample collection, radioubsentiatry, and counting.

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